

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
15 March 2001 (15.03.2001)

PCT

(10) International Publication Number
WO 01/18145 A2

(51) International Patent Classification⁷: C09K 3/30

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(21) International Application Number: PCT/GB00/03426

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(22) International Filing Date:
5 September 2000 (05.09.2000)

(81) Designated States (national): AE, AG, AL, AM, AT, AU,
AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ,
DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR,
HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR,
LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ,
NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM,
TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
9921037.9 7 September 1999 (07.09.1999) GB

(84) Designated States (regional): ARIPO patent (GH, GM,
KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian
patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European
patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE,
IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG,
CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

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Published:

— Without international search report and to be republished
upon receipt of that report.

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

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WO 01/18145 A2

(54) Title: ELECTROSTATIC AEROSOL COMPOSITIONS

(57) Abstract: An electrically neutral composition in the form of a water-in-oil or an oil-in-water emulsion is imparted with a unipolar electrostatic charge on spraying from an aerosol spray device by incorporating into the composition at least one polar or ionic or aromatic or linearly conjugated compound. The amount of the polar or ionic or aromatic or linearly conjugated compound included in the composition is from 0.01 to 80% w/w based on a non-ionic surfactant also included in the composition, but is such that the theoretical conductivity of the emulsion is less than the bulk conductivity of the emulsion.

ELECTROSTATIC AEROSOL COMPOSITIONS

The present invention relates to aerosol compositions and, in particular, compositions in which the droplets are imparted with an electrostatic charge on spraying from an aerosol spray device and in which the electrostatic charge on the droplets is maximised through the inclusion in the compositions of certain selected components.

Aerosol spray devices are a convenient form in which a variety of useful products, such as insecticides, air fresheners, antiperspirants, hair sprays, horticultural products, waxes and polishes, oven cleaners, starches and fabric finishes, shoe and leather care products, glass cleaners and various other household, institutional, professional or industrial products, can be dispensed.

The utility of aerosol spray devices resides in the ability to readily deliver the composition contained within the device in the form of fine droplets to the target area, for example the spraying of an insecticide onto target insects.

In WO 97/28883 there is described a method of precipitating airborne particles from air in a domestic environment containing such particles in which the air to be treated is sprayed with liquid droplets from an aerosol spray device with a unipolar charge being imparted to the droplets during the spraying of the liquid droplets by the aerosol spray device, the unipolar charge being at a level such that the droplets have a charge to mass ratio of at least $\pm 1 \times 10^{-4}$ C/kg.

In WO 99/01227 there is described a method of killing flying insects by spraying into the air in which the insects are flying liquid droplets of an

insecticidal composition, a unipolar charge being imparted to the liquid droplets by double layer charging and charge separation during spraying, the unipolar charge being at a level such that the said liquid droplets have a charge to mass ratio of at least $+/- 1 \times 10^{-4}$ C/kg. An apparatus for imparting the unipolar charge of this magnitude to a liquid composition is also described.

We have now found that by careful selection of the components which are to be contained within a liquid composition for application by aerosol spraying, it is possible to charge the liquid droplets during the spraying operation without requiring any special features of the construction of the aerosol spraying head.

Accordingly, in one aspect the present invention provides an electrically neutral composition in the form of a water-in-oil or an oil-in-water emulsion, in which droplets of the emulsion on discharge from an aerosol spray device are imparted with a unipolar electrostatic charge, which composition comprises:

- (a) at least one propellant in an amount of from 2 to 80% w/w;
- (b) at least one non-ionic surfactant in an amount of from 0.01 to 10% w/w;
- (c) optionally one or more oils or solvents, preferably aliphatic, linearly conjugated or aromatic, within the oil phase in an amount of up to 80% w/w, preferably up to 40% w/w;
- (d) at least one polar or ionic or aromatic or conjugated compound in an amount of from 0.01 to 80% w/w based on the non-ionic surfactant, but which is such that the theoretical conductivity of the emulsion is less than the bulk conductivity of the emulsion; and

(e) water.

In a second aspect the present invention provides a method of enhancing the unipolar charge which is imparted to droplets of an emulsion on discharge from an aerosol spray device in which the droplets are formed from an oil-in-water or a water-in-oil emulsion composition which comprises: .

(a) at least one propellant in an amount of from 2 to 80% w/w;

(b) at least one non-ionic surfactant in an amount of from 0.01 to 10% w/w;

(c) optionally one or more oils or solvents, preferably aliphatic, linearly conjugated or aromatic, within the oil phase in an amount of up to 80% w/w. preferably up to 40% w/w;

(d) at least one polar or ionic or aromatic or linearly conjugated compound in an amount of from 0.1 to 80% w/w based on the non-ionic surfactant, but which is such that the theoretical conductivity of the emulsion is less than the bulk conductivity of the emulsion; and

(e) water.

In a third aspect the present invention provides the use of a non-ionic surfactant and at least one polar or ionic or

aromatic or conjugated compound in an amount of from 0.01 to 80% w/w based on the non-ionic surfactant to enhance the electrostatic charge imparted to droplets of a composition in the form of a water-in-oil or an oil-in-water emulsion on discharge from an aerosol spray device, which composition includes:

(a) at least one propellant in an amount of from 2 to 80% w/w;

(b) optionally one or more oils or solvents, preferably aliphatic, linearly conjugated or aromatic,

within the oil phase in an amount of up to 80% w/w, preferably up to 40% w/w; and

(c) water;

and the amount of the polar or ionic or aromatic or conjugated compound being such that the theoretical conductivity of the emulsion is less than the bulk conductivity of the emulsion.

In a fourth aspect the present invention provides an aerosol spray which contains an electrically neutral composition in the form of a water-in-oil emulsion, an oil-in-water emulsion or a single phase composition, in which droplets of the composition on discharge from the aerosol spray device are imparted with a unipolar electrostatic charge, wherein the formulation of the composition and the material of the portion of the aerosol spray device with which the liquid comes into contact on spraying are selected such that

- i) the difference between the Lewis base component of the liquid and the Lewis base component of the material with which the liquid comes into contact on spraying is at least $\pm 2 \text{mJ m}^{-2}$;
and/or;
- ii) the difference between the Lewis acid component of the liquid and the Lewis acid component of the material with which the liquid comes into contact on spraying is at least $\pm 0.5 \text{ mJ m}^{-2}$.

The liquid droplets preferably have a charge to mass ratio of at least $\pm 1 \times 10^{-4} \text{ C/kg}$, more preferably at least $\pm 2 \times 10^{-4} \text{ C/kg}$. The higher the charge to mass ratio of the liquid droplets, the more effective the liquid droplets will be for their intended use, such as precipitating airborne particles

and targeting insects. This charge level is considerably higher than the charge level which is achieved when spraying conventional liquid formulations from conventional aerosol spray device where charge levels of the order of $+/- 1 \times 10^{-5}$ to $+/- 1 \times 10^{-8}$ C/kg are obtained.

In the formulations of the present invention it is the combination of components (b) and (d) of the emulsion which improves the electron transfer through the emulsion with the charge being transferred from droplet to droplet through the emulsion at the interface between the disperse phase and the continuous phase.

The various components (a) to (e) of the compositions of the present invention are discussed in turn below:

Propellants

One or more propellants are used in the composition of the invention in a total amount of from 2 to 80% w/w. Amongst the propellants that may be used are hydrocarbons and compressed gas, of which hydrocarbons are preferred.

Hydrocarbon propellants which may be used are acetylene, methane, ethane, ethylene, propane, n-butane, n-butene, isobutane, isobutene, pentane, pentene, isopentane and isopentene. Mixtures of these propellants may also be used. Commercially available propellants typically contain a number of hydrocarbon gases. For example, an odourised commercial butane, contains predominantly n-butane and some iso-butane together with small amounts of propane, propene, pentane and butene.

Preferred hydrocarbon propellants include propane, n-butane, isobutane, pentane and isopentane,

whilst the most preferred are propane, iso-butane and n-butane.

Particularly preferred hydrocarbon propellants are mixtures of propane, n-butane and iso-butane.

Whilst broadly the concentration of hydrocarbon propellant will be from 2 to 80% w/w, generally the concentration will be from 10 to 60% w/w, preferably 25 to 60% w/w and most preferably about 40% w/w.

When compressed gases are used as a propellant these will generally be carbon dioxide, nitrogen or air. Usually, they will be used at a concentration of 2 to 20% w/w, preferably about 5% w/w.

Non-ionic Surfactants

Non-ionic surfactants for use in the present invention include mono, di and tri sorbitan esters, polyoxyethylene mono, di and tri sorbitan esters; mono and polyglyceryl esters; alkoxylated alcohols; alkoxylated amines; alkoxylated acids; amine oxides; ethoxylated/proxyolated block copolymers; alkoxylated alkanolamides; and alkoxylated alkyl phenols.

Particularly preferred are those surfactants which contain at least one alkyl, allyl or substituted phenyl group containing at least one C₆ to C₂₂ carbon chain. Examples are esters with C₁₀-C₂₂ fatty acids, preferably C₁₂-C₁₈ fatty acids, particularly polyglycerol oleate and ethoxylated fatty alcohols, such as oleyl alcohol ethoxylated with two moles of ethylene oxide. Further examples are the polyethylene glycol oleates, such as PEG-4 oleate, PEG-8 oleate and PEG-12 oleate.

In some instances, the non-ionic surfactant may itself be combined with component (d). For example, when the non-ionic surfactant is polyglycerol oleate,

the surfactant may contain small quantities of sodium or potassium oleates as impurities as a by-product of manufacture. For example is an amount of from 0.01 to 1% by weight. Greater quantities of such ionic compounds are generally not desirable and may result in the compositions not fulfilling the conductivity requirements of the compositions of the invention.

The concentration of the non-ionic surfactant is from 0.01 to 10% w/w, preferably 0.01 to 1% w/w.

Polar, ionic, aromatic or conjugated compounds

The polar or ionic or aromatic or conjugated compound which is included as component (d) in the compositions of the present invention is preferably a compound which is attracted to the interface between the disperse phase and the continuous phase and may be selected from:

a) alkali metal salts, alkaline earth metal salts, ammonium salts, amine salts or amino alcohol salts of one or more of the following compounds: alkyl sulphates, alkyl ether sulphates, alkylamidoether sulphates, alkylarylpolyether sulphates, monoglyceride sulphates, polyglyceride sulphates, alkyl sulphonates, alkylamine sulphonates, alkyl-aryl sulphonates, olefin sulphonates, paraffin sulphonates, alkyl sulpho-succinates, alkylether sulphosuccinates, alkylamide sulphosuccinates, alkyl sulphocinnamates, alkyl sulphoacetates, alkyl phosphates, alkylether phosphates, acyl sarcosinates, acyl isothionates and N-acyl taurates;

b) alkyl amidopropylbetaines, alkylamido-betaines, alkylamidosulphobetaines, alkylbetaines, aminimides, quaternary ammonium compounds and quaternary phosphonium compounds;

c) carboxylic acids, carboxylic acid salts, esters, ketones, aldehydes, amides or amines of carboxylic acids containing from 6 to 30 carbon atoms;

d) diethyl orthophthalate, methylphenylcarbonyl acetate, α -methyl ionone, 4-hydroxy 3-methoxy-benzaldehyde, phenylethyl alcohol, dipropylene glycol, styryl acetate, n-butyl benzoate, isopropyl 4-hydroxybenzoate, isobutyl acetophenone, isopropyl acetophenone, nicotinic acid, benzoic acid, 2-naphthol, neopentyl benzene, naphthalene, toluene, fullerene, tannic acid, t-butylacetophenone, isopropylcinnamate, resorcinol, 4-methoxycinnamaldehyde, arbutin, 4-acetoxy-3-methoxycinnamaldehyde, 4-isopropylphenol, trans-stilbene, esculetin, p-chloro-m-xylenol, chloro-o-cresol, triclosan, norfenefrine, norepinephrine, hexyl-resorcinol, limonene, methylphenylcarbonyl acetate and p-tert-butyl- α -methylhydrocinnamic aldehyde.

Particularly preferred compounds in group (b) are alkyldimethylbenzyl ammonium chloride, octyltrimethyl ammonium bromide, cetyltrimethylammonium bromide and dodecyltrimethylphosphonium bromide.

Particularly preferred compounds in group (c) are lauric, oleic, palmitic, ricinoleic and stearic acids, or the salts, amides, esters, ketones or aldehydes thereof.

It will be understood that certain of the aromatic or conjugated compounds may also be classed as fragrances.

The concentration of component (d) is from 0.01 to 80% w/w, preferably from 0.01 to 30%, more preferably from 0.01 to 10% w/w based on the non-ionic surfactant, component (b). The amount of component (d) is selected so that the bulk conductivity is

greater than the theoretical conductivity. In some cases, too great an amount of component (d) can result in the composition not fulfilling the conductivity requirement of the compositions of the invention.

It will be understood that mixtures of compounds may be used as component (d). In particular it has been found that the addition of an aromatic compound together with an ionic compound increases the charge to mass ratio of the formulations on spraying.

Oils or Solvents

One or more oils or solvents may be incorporated in the compositions of the invention in an amount of up to 80% w/w, preferably up to 40% w/w. Generally, the solvent will be water immiscible.

A wide range of oils or solvent materials may be used, although care should be exercised to ensure that the solvent does not adversely interact with any active components of the compositions of the invention, such as insecticides.

Examples of solvents that may be used in the compositions of the invention include:-

liquid n-paraffins, liquid isoparaffins, cycloalkanes, naphthene-containing solvents, white spirit, kerosene, ester solvents, silicone solvents or oils, fatty acids, dialkyl phthalates, C₅-C₁₁ alcohols and fatty alcohols. Specific examples of these are as follows:-

liquid n-paraffins - Norpar 12, Norpar 13 and Norpar 15 (available from Exxon)

liquid isoparaffins - Isopar G, Isopar H, Isopar L, Isopar M and Isopar V (available from Exxon).

Naphthene-containing solvents - Exxsol D40, Exxsol D60, Exxsol D80, Exxsol D100, Exxsol D110,

Nappar 10, Solvesol 100, Solvesol 150, Solvesol 200 (available from Exxon)

Ester solvents - such as alkyl acetates, examples being Exxate 1000, Exxate 1300 (available from Exxon), and Coasol (available from Chemoxy International);

Silicone solvent oils - Dow Corning 244, 245, 344 and 345 fluids,

Fatty alcohols - octanol, dodecanol, lauryl alcohol, myristyl alcohol, cetyl alcohol, stearyl alcohol, cetostearyl alcohol, oleyl alcohol.

Preferred solvents are liquid hydrocarbon solvents, n-paraffins, and iso-paraffins.

Although the solvent is preferably incorporated at a level of from 1 to 20% w/w, more preferably the concentration will be in the range of from 2 to 10% w/w, most preferably about 5% w/w.

Insecticidal Compositions

In one preferred agent of the present invention the compositions are insecticidal compositions which contain from 0.001 to 5% w/w of an insecticidal compound. A wide range of active ingredients may be used of which pyrethroids, particularly synthetic pyrethroids, chlorpyrifos, propoxur and diazinon are preferred.

When synthetic pyrethroids such as lambda cyhalothrin and bioresmethrin are used, generally they will be incorporated in concentrations of about 0.02% w/w or above.

Other synthetic pyrethroids such as cypermethrin, tetramethrin, permethrin and bioallethrin, will usually be incorporated to give a concentration of about 0.2%-0.5% w/w, or above.

Chloropyrifos, propoxur and diazinon will generally be incorporated to give concentration in the range of 0.5-0.9% w/w.

Preferably, insecticidal compositions of the invention will include an insecticide which functions primarily to knock down an insect, together with a second insecticide which functions primarily as a kill agent. An example of such a combination is the use of permethrin as a kill agent in a combination with tetramethrin as a knock down agent.

Optional Ingredients

Various optional ingredients may be incorporated into the compositions of the present invention. For example, in order to maximise the effectiveness of the insecticidal activity of the compositions of the invention, synergists such as N-octylbicycloheptene dicarboximide and piperonyl butoxide may be included at a concentration of from 0.5 to 1.5% w/w, most preferably about 1.0% w/w, for use in conjunction with pyrethroid insecticides.

In addition, other ingredients including corrosion inhibitors, such as 1-hydroxyethyl-2-heptadecenyl imidazoline and/or sodium benzoate, preferably in a concentration from 0.01 to 0.5% w/w, preservatives and antioxidants, such as butylated hydroxytoluene, may be used as required. One or more fragrance components may also be included, according to the particular consumer requirements. It will be understood that certain fragrance components are components which may comprise component (d) of the compositions of the present invention and in this instance, such a component is not an optional ingredient.

Lewis Acid and Lewis Base Characteristics

In relation to the fourth embodiment of the invention an aerosol spray device and the electrically neutral composition contained therein have certain Lewis acid and Lewis base characteristics which assist in imparting a unipolar charge to the liquid.

When two substances are brought together and then separated, an electrical charge is transferred from one to the other. This can occur for solid-solid separations, for solid-liquid separations and for liquid-liquid separations. When one of the components becomes airborne, the electrical charge can remain on the substance for a significant length of time as there is no place to ground the charge. The ability of substances to transfer their charge can be related to the characteristic Lewis acid (γ^+) and Lewis base (γ^-) values for the substance. All substances have characteristic values and these can be calculated indirectly from their component surface energies.

These surface energies when combined with a London-van der Waals component (γ^{LW}) form what is known as the surface tension. This is easiest to measure at the interface between a drop of liquid on a solid substrate.

This equation used for these calculations comes from the approach used by Good and van Oss:

$$(1 + \cos \theta) \gamma_L = 2[\sqrt{(\gamma_s^{LW} \gamma_L^{LW})} + \sqrt{(\gamma_s^+ \gamma_L^-)} + \sqrt{(\gamma_s^- \gamma_L^+)}]$$

Where θ is the contact angle that a drop of liquid makes with the surface.

γ_s is the energy component for the solid.

γ_L is the energy component for the liquid.

γ^{LW} Represents the London-van der Waals component.

γ^+ is the Lewis acid component (electron acceptor).

γ^- is the Lewis base component (electron donor).

Using three or more test liquids whose characteristics are known, it is possible to solve this equation for the three unknowns, γ_s^{LW} , γ_s^+ and γ_s^- . By using test solids whose characteristics are known, it is also possible to solve this equation to find the three unknown liquid surface energies. Thus it is possible to characterise a series of solids and liquids to form a series of γ^+ and γ^- values. In the case of solids this series matches that of the triboelectric series. Below is a non-exclusive list of solid substances that form part of this triboelectric series. The values are scaled such that water has a γ^- and γ^+ of 25 mJ m⁻². The top of the list tends to become positive, when separated from a substance at the bottom of the list, which would become negative.

Surface	γ^- (mJ m ⁻²)
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+ Glass	16
Acetal 900P NC-10	15
Celluloid	13.8
PMMA	12
Nylon	11.3
PVC	8.4
Polyester 2002-2	5
Polyethylene	0.1
Polypropylene	0.04
- PTFE	0

Liquids also have a value of γ^- and γ^+ that can be measured. When liquids contact a solid of known γ^- and γ^+ on aerosol spraying, and the γ^+ of the liquid is greater than, or less than that of the solid by 2 mJ m^{-2} , preferably by 5 mJ m^{-2} , most preferably 15 mJ m^{-2} , or a γ^+ greater than, or less than that of the solid by 0.5 mJ m^{-2} preferably 1 mJ m^{-2} , most preferably 2 mJ m^{-2} , the liquid has a tendency to charge to $+/- 1 \times 10^{-4} \text{ C/kg}$. A common material from which inserts for aerosol actuators are made is Acetal 900P NC-10, which has a γ^- of 15 mJ m^{-2} .

Aerosol Spray Devices

The compositions of the present invention, when sprayed through conventional aerosol spray heads, form into droplets which are imparted with a unipolar charge of at least $+/- 1 \times 10^{-4} \text{ C/kg}$.

It is possible to impart even higher charges to the liquid droplets by choosing aspects of the aerosol device including the material, shape and dimensions of the actuator, the actuator insert, the valve and the dip tube and the characteristics of the liquid which is to be sprayed, so that the required level of charge

is generated as the liquid is dispersed as droplets. A number of characteristics of the aerosol system increase double layer charging and charge exchange between the liquid formulation and the surface of the aerosol system. Such increases are brought about by factors which may increase the turbulence of the flow through the system, and increase the frequency and velocity of contact between the liquid and the internal surface of the container and valve and actuator system.

By way of example, characteristics of the actuator can be optimised to increase the charge levels on the liquid sprayed from the container. A smaller orifice in the actuator insert, of a size of 0.45mm or less, increases the charge levels of the liquid sprayed through the actuator. The choice of material for the actuator can also increase the charge levels on the liquid sprayed from the device with materials such as nylon, polyester, acetal, PVC and polypropylene tending to increase the charge levels. The geometry of the orifice in the insert can be optimised to increase the charge levels on the liquid as it is sprayed through the actuator. Inserts which promote the mechanical break-up of the liquid give better charging.

The actuator insert of the spray device may be formed from a conducting, insulating, semi-conducting or static-dissipative material.

The characteristics of the dip tube can be optimised to increase charge levels in the liquid sprayed from the container. A narrow dip tube, of for example about 1.27 mm internal diameter, increases the charge levels on the liquid, and the dip tube material can also be changed to increase charge.

Valve characteristics can be selected which increase the charge to mass ratio of the liquid product as it is sprayed from the container. A small tailpiece orifice in the housing, of about 0.65 mm, increases the charge to mass ratio during spraying. A reduced number of holes in the stem, for example 2 x 0.50mm, also increases the charge during spray. The presence of a vapour phase tap helps to maximise the charge levels, a larger orifice vapour phase tap of, for example, about 0.50 mm to 1.0 mm generally giving higher charge levels.

The liquid droplets sprayed from the aerosol spray device will generally have diameters in the range of from 5 to 100 micrometres, with a peak of droplets of about 40 micrometres.

Preparation

The compositions of the present invention may be prepared by standard techniques which are well known in the art. For example, components (b) to (d) may be mixed together to form the solvent phase. This solvent phase is then mixed with water to produce an emulsified concentrate which is then filled into cans and blended with the propellant. Alternatively, the concentrate and the propellant may be filled into the cans simultaneously.

Conductivity

The theoretical conductivity of an emulsion, σ , can be calculated from measurements of the actual conductivity of the external phase and the internal phase, according to the following equation:

$$\sigma = \sigma_c(1 + 3\varphi(\sigma_p - \sigma_c)/(\sigma_p + 2\sigma_c))$$

where σ = theoretical conductivity of the emulsion

σ_c = measured conductivity of the separated external phase
 σ_p = measured conductivity of the separated internal phase
 φ = volume fraction of the internal phase.

The bulk conductivity can be determined by experimentation. Component (d) of the compositions of the present invention serves to enhance the actual conductivity of the emulsion and thus the bulk conductivity of the emulsion is higher than the theoretical conductivity calculated according to the above formula. Preferably the difference between the theoretical conductivity of the emulsion of the emulsion is at least + 0.5 $\mu\text{S cm}^{-1}$, preferably at least 74 $\mu\text{S cm}^{-1}$, more preferably at least + 6 $\mu\text{S cm}^{-1}$. Component (d) in the compositions of the present invention thus improves the electron transfer through the emulsion with the charge being transferred from droplet to droplet through the emulsion at the interface between the disperse phase and the continuous phase.

The present invention will be further described with reference to the following non-limiting Examples.

Method for predicting the theoretical conductivity of an emulsion through the measurement of the conductivity of the individual phases:

1. Calibrate the conductivity cell by measuring a solution of known conductivity. The conductivity cell comprises a pair of platinum electrodes, held apart and attached to the inside of a glass tube of approximately 1 cm internal diameter.

2. Using the same cell, measure the conductivity of the bulk emulsion according to the invention, whilst ensuring that the emulsion is static and homogeneous prior to taking the measurement.
3. Determine whether the continuous phase of the emulsion is water or oil.
4. Separate the two phases of the emulsion by either gravimetric or centrifugal separation. Isolate the phases and measure the conductivity of each phase in the calibrated cell.
5. Use the equation given below to determine the theoretical conductivity of the bulk emulsion.
6. The difference between the theoretical conductivity and that obtained directly from the measurements is the contribution to the bulk conductivity due to the emulsion droplets.

$$\sigma = \sigma_c(1 + 3\psi(\sigma_p - \sigma_c) / (\sigma_p + 2\sigma_e))$$

where σ = theoretical conductivity of the emulsion

σ_c = measured conductivity of the separated external phase

σ_p = measured conductivity of the separated internal phase

ψ = volume fraction of the internal phase.

Measurement of Electrostatic Charge

The charge to mass ratio of the compositions of Examples 4 to 50 was measured using a standard aerosol can with a valve insert made from polyoxymethylene

according to the design shown in accompanying Figure 1 in which

Fig. 1a is a plan view;

Fig. 1b is a sectional view; and

Fig. 1c is a inverted plan view of the valve insert.

EXAMPLE 1 (Comparative)

An emulsion was prepared from the following ingredients:

<u>Ingredient</u>	<u>%</u>
Ethoxylated (7EO) alcohol (C ₁₂ -C ₁₅)	0.24 w/v
Deionised water	47 v/v
Decane	55 v/v
Actual (measured) conductivity of the bulk emulsion	7.4 μ S cm ⁻¹
σ_c	16.6 μ S cm ⁻¹
σ_p	4.1 μ S cm ⁻¹
Assuming φ =	0.5
σ	8.2 μ S cm ⁻¹

Difference (conductivity due the emulsion droplets) = 7.4 - 8.2 = -0.8 μ S cm⁻¹.

This composition did not contain any component (d) and as can be seen the bulk conductivity of the emulsion is less than the theoretical value.

EXAMPLE 2

An emulsion was prepared from the following ingredients:

<u>Ingredient</u>	<u>%</u>
Ethoxylated (7EO) alcohol (C ₁₂ -C ₁₅)	0.24 w/v
Sodium lauryl sulphate	3% w/w of the

(30% active)	non-ionic surfactant
Deionised water	47 v/v
Decane	53 v/v
Actual (measured) conductivity of the bulk emulsion	$22.3 \mu\text{S cm}^{-1}$
σ_c	$39.4 \mu\text{S cm}^{-1}$
σ_p	$4.0 \mu\text{S cm}^{-1}$
Assuming $\varphi =$	0.5
σ	$14.1 \mu\text{S cm}^{-1}$
Difference (conductivity due the emulsion droplets) = $22.3 - 14.1 = 8.2 \mu\text{S cm}^{-1}$.	

EXAMPLE 3 (Comparative)

An emulsion was prepared from the following ingredients:

<u>Ingredient</u>	<u>%</u>
Ethoxylated (7EO) alcohol (C ₁₂ -C ₁₅)	0.24 w/v
Sodium lauryl sulphate (30% active)	6% w/w of the non-ionic surfactant
Deionised water	47 v/v
Decane	53 v/v
Actual (measured) conductivity of the bulk emulsion	$23.3 \mu\text{S cm}^{-1}$
σ_c	$83.0 \mu\text{S cm}^{-1}$
σ_p	$9.0 \mu\text{S cm}^{-1}$
Assuming $\varphi =$	0.5
σ	$30.3 \mu\text{S cm}^{-1}$
Difference (conductivity due the emulsion droplets) = $23.3 - 30.3 = -7.0 \mu\text{S cm}^{-1}$.	

This composition contains a large amount of component (d) and as can be seen the bulk conductivity of the emulsion is less than the theoretical value.

EXAMPLE 4

<u>Ingredient</u>	<u>%</u>
Ethoxylated (SEO) alcohol	
(C ₁₂₋₁₅) incorporating benzalkonium	
chloride (50% active) at 2% w/w in	
surfactant	0.24% w/v
Deionised water	47% v/v
Butane 40	53% v/v

This formulation when made up as an aerosol and sprayed though the physical valve/actuator combination described above produced a mono-polar charge on the sprayed droplets of $+1.65 \times 10^{-4}$ C/Kg.

The same formulation was prepared substituting decane for butane in order that the conductivity could be measured.

Actual (measured) conductivity	
of the bulk emulsion	$15.7 \mu\text{S cm}^{-1}$
σ_c	$40.2 \mu\text{S cm}^{-1}$
σ_p	$3.3 \mu\text{S cm}^{-1}$
Assuming $\theta =$	0.5
σ	$13.7 \mu\text{S cm}^{-1}$
Difference (conductivity due to the	
emulsion droplets) = $15.7 - 13.7 = 2.0 \mu\text{S cm}^{-1}$	

EXAMPLE 5Formulation 1

<u>Ingredient</u>	<u>% w/w</u>
Solvent	5.0
Fragrance component	0.70
Butylated hydroxytoluene	0.013
Polyglyceryl oleate	0.30
Deionised Water	58.99
Butane 40	35

The solvent used and the fragrance component that can be used in the above formulation are illustrated in the following examples:

Example	Solvent	Fragrance Component	Charge/Mass ($\times 10^{-4}$ C/kg)
5	Isopar E	diethyl-o-phthalate	-2.2
6	Isopar E	styralyl acetate	-2.5
7	Isopar G	α -methyl ionone	-1.9
8	Isopar G	vanillin	-1.6
9	heptane	Litsea Cybaba	-1.7
10	pentane	Lilial	-2.3
11	Isopar E	phenylethyl alcohol	-2.4
12	Isopar L	dipropylene glycol	-2.2

The Isopar E, G and L range of solvents can be obtained from Exxon.
 The fragrance components used were obtained from Robertet Ltd.

EXAMPLE 13Formulation 2

<u>Ingredient</u>	<u>% w/w</u>
Solvent	5.0
Aromatic component	0.70
Butylated hydroxytoluene	0.013
Polyglyceryl oleate	0.30
Deionised Water	58.99
Butane 40	35

The solvent used and the aromatic component can be used in the above formulation are illustrated in the following examples:

Example	Solvent	Aromatic Component	Charge/Mass (x10 ⁻⁴ C/kg)
13	Solvesol 150	Cleanox fragrance	-3.0
14	Isopar E	n-butyl benzoate	-2.5
15	Isopar L	isopropyl-4-hydroxybenzoate	-3.0
16	Isopar E	isobutyl acetophenone	+1.6
17	heptane	isopropyl acetophenone	+1.7
18	pentane	benzoic acid	+1.2
19	Isopar V	2-naphthol	-5.3
20	Isopar G	toluene	+1.9
21	Pentane	neopentyl benzene	-5.9
22	Isopar C	naphthalene	-5.4
23	Isopar G	fullerene C60/70	-4.5

EXAMPLE 24Formulation 3Ingredient

<u>Ingredient</u>	<u>%</u>
Ethoxylated (5EO) alcohol (C ₁₂₋₁₅) incorporating sodium laureth sulphate (30% active)	
at 4% w/w in surfactant	0.24% w/v
Deionised water	47% v/v
Butane 40	53% v/v

This formulation, when made up as an aerosol and sprayed through the physical valve/actuator combination described above produced a mono-polar charge on the sprayed droplets of -1.1×10^{-4} C/kg.

The same formulation was prepared substituting decane for butane. The formulation had a γ' of 26.2 mJ m⁻².

An acetal 900P NC-10 insert in the spray head had a γ' of 15 mJ m⁻².

The difference between these Lewis base values = 26.2 - 15 = 11.2 mJ m⁻².

Ingredient	%w/w	Example 25	%w/w	Example 26	%w/w	Example 27	%w/w	Example 28	%w/w
Bioallethrin	0.194	0.914	0.194	0.036	0.036	0.194	0.036	0.194	0.194
Bioresmethrin	0.036	0.036	0.02	0.02	0.02	0.02	0.02	0.02	0.02
BHT	0.02	0.900	0.900	0.900	0.900	0.900	0.900	0.900	0.900
Polyglycerol oleate	0.900	0.09	0.180	0.180	0.100	0.100	0.100	0.100	0.100
Oleic acid	0.09	0.100	0.100	0.100	0.100	0.100	0.100	0.100	0.100
Perfume	7.500	7.500	7.500	7.500	7.500	7.500	7.500	7.500	7.500
Norpar 13	51.16	51.07	51.07	40.000	40.000	51.205	40.000	40.000	51.227
Deionised water									
H55									
Charge/mass(x10 ⁻⁴ C/kg)	-0.75	-0.72	-0.72	-0.63	-0.63	-0.63	-0.63	-0.63	-0.90
Ingredient	%w/w	Example 29	%w/w	Example 30	%w/w	Example 31	%w/w	Example 32	%w/w
Teric 12A2	0.800	1.000	1.000	0.400	0.400	0.800	0.400	0.400	0.700
Oleic acid	0.400	7.500	7.500	51.300	51.100	7.500	51.300	51.400	0.400
Norpar 13	40.000	40.000	40.000	40.000	40.000	40.000	40.000	40.000	7.500
Deionised water									
H55									
Charge/mass(x10 ⁻⁴ C/kg)	-1.02	-0.68	-0.68	-0.68	-0.68	-0.816	-0.816	-0.816	-0.816

Ingredient	%w/w	%w/w	%w/w	Example 33	Example 34	Example 35
Teric 12A2	0.600	0.500	1.000			
Oleic acid	0.400	0.400	0.200			
Norpar 13	7.500	7.500	7.500			
Deionised water	51.500	51.600	51.300			
H55	40.000	40.000	40.000			
Charge/mass (x10 ⁻⁴ C/kg)	-1.596	-0.966	-1.53			

Ingredient	%w/w	%w/w	%w/w	Example 36	Example 37
Teric 12A2	1.000	1.000	1.000		
Oleic acid	0.400	0.400	0.800		
Perfume	0.100	0.100	0.100		
Norpar 13	7.500	7.500	7.500		
Deionised water	51.100	51.700	50.700		
H55	40.000	40.000	40.000		
Charge/mass (x10 ⁻⁴ C/kg)	-0.57	-0.738	-0.738		

Ingredient	%w/w	%w/w	%w/w	Example 40
	Example 38	Example 39	Example 41	Example 42
Teric 12A2	1.000	1.000	1.000	1.000
Lauric acid	0.100	0.200	0.010	0.020
Norpar 13	7.500	7.500	7.500	7.500
Deionised water	51.400	51.300	51.300	51.300
H55	40.000	40.000	40.000	40.000
Charge/mass (x10 ⁻⁴ C/kg)	-0.532	-0.578	-0.502	-0.704

Ingredient	%w/w	%w/w	%w/w	Example 41	Example 42
	Example 38	Example 39	Example 40	Example 41	Example 42
Teric 12A2	1.000	1.000	1.000	1.000	1.000
Palmitic acid	0.010	0.020	0.010	0.020	0.020
Norpar 13	7.500	7.500	7.500	7.500	7.500
Deionised water	51.400	51.300	51.300	51.300	51.300
H55	40.000	40.000	40.000	40.000	40.000
Charge/mass (x10 ⁻⁴ C/kg)	-0.532	-0.578	-0.502	-0.502	-0.704

Ingredient	%w/w	Example 43	%w/w	Example 44	%w/w	Example 45	%w/w	Example 46	%w/w
Teric 12A2	0.700	0.720	0.700	0.700	0.700	0.700	0.700	0.700	0.700
Oleic acid	0.500	0.400	0.400	0.400	0.400	0.400	0.400	0.400	0.400
Norpar 13	7.500	7.500	7.500	7.500	7.500	7.500	7.500	7.500	7.500
Deionised water	41.300	31.38	51.400	51.400	51.300	51.300	51.300	51.300	51.300
H55	-	-	-	-	-	-	-	-	40.000
H46	50.000	60.000	40.000	40.000	-	-	-	-	-
Charge/mass (x10 ⁻⁴ C/kg)	-1.39	-2.12	-0.71	-0.71	-1.65	-1.65	-1.65	-1.65	-1.65

Ingredient	%w/w	Example 47
Teric 17A2	0.85	
Oleic acid	0.35	
Norpar 13	5.00	
Deionised water	33.80	
H46	60.000	
Charge/mass (x10 ⁻⁴ C/kg)	-4.8	

Ingredient	%w/w	Example 48	%w/w	Example 49
Norpar 13	5.00	5.00	5.00	
Bioallethrin	0.25	0.25	0.25	
Bioresmethrin	0.05	0.05	0.05	
BHT	0.02	0.02	0.02	
Deionised water	33.28	33.28	33.58	
Teric 17A3	0.60	0.60	0.35	
Crill 45	0.40	0.40	0.35	
Pine Fragrance	0.10	0.10	0.10	
Oleic acid	0.30	0.30	0.30	
H46	60.00	60.00	60.00	
Charge/mass (x10 ⁻⁴ C/kg)	-1.41	-1.41	-1.34	

Ingredient	%w/w	Example 50
Bioallethrin	0.209	
Bioresmethrin	0.039	
BHT	0.005	
Polyglycerol oleate (containing from 0.01 to 1% by weight of sodium or potassium oleate)	0.900	
Perfume	0.100	
Norpar 13	7.500	
Deionised water	51.247	
H55	40.000	
Charge/mass (x10 ⁻⁴ C/kg)	-1.59	

Ingredient	Comparative	Comparative	Comparative
	Example A	Example B	Example C
Teric 12A2	-	1.000	1.000
Bioallethrin	0.194	-	-
Bioresmethrin	0.036	-	-
BHT	0.02	-	-
Polyglycerol oleate	0.900	-	-
Perfume	0.100	-	-
Norpar 13	7.500	7.500	7.500
Deionised water	51.25	51.50	51.500
H55	40.000	40.000	40.000
Crill 45	0.40	0.35	-0.017
Charge/mass (x10 ⁻⁴ C/kg)	-0.35	+0.21	-0.017

EXAMPLES 51 to 61

The following Table provides details of compositions in which the amount of oleic acid, sodium oleate or a mixture of oleic acid and sodium oleate contained in the compositions is varied.

These formulations were sprayed through the physical value/activator combination described above and the monopolar charges on the sprayed droplets were recorded.

The Lewis acid and base values for the compositions were also recorded. The Lewis base value of the Acetal 900P NC-10 insert material through which the compositions were sprayed was 15mJ m^{-2} . The Lewis acid value of the Acetal 900P NC-10 insert material through which the compositions were sprayed was 0mJ m^{-2}

Values are also given for the theoretical conductivity and bulk conductivity of each of the formulations.

Example No Components & w/w	51	52	53	54	55
Deionized water	59.0759	59.0730	59.0585	59.0730	59.0585
Fragrance Cleanox	0.2470	0.2470	0.2470	0.2470	0.2470
Isopar G	4.9985	4.9985	4.9985	4.9985	4.9985
Columed MBQ Crestor	0.5785	0.5785	0.5785	0.5785	0.5785
L(PEO) - Croda	-	0.0029	0.0174	-	-
Na Oleate	-	-	-	0.0029	0.0174
Oleic Acid	-	-	-	0.0029	0.0174
BHT 0.1001	0.1001	0.1001	0.1001	0.1001	-
Butane 40	5.0000	35.0000	35.0000	35.0000	35.0000
Charge/Mass ($\times 10^{-4}$ C/Kg)	-2.08	-2.05	-0.72	-1.64	-1.40
Lewis base value	19.2	12.0	17.0	26.0	17.5
γ^- (mJ m^{-2})					
Lewis acid value	2.0	3.0	1.0	1.0	2.5
γ^+ (mJ m^{-2})					
Theoretical	-	-	3.87	2.63	2.47
Conductivity ($\mu\text{S cm}^{-1}$)					
Bulk Conductivity ($\mu\text{S cm}^{-1}$)	-	-	4.55	3.33	2.63

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Example No Components & w/w	56	57	58	59	60	61
Deionized water	59.0747	59.0701	59.0641	59.0411	59.0556	59.0556
Fragrance Cleanox	0.2470	0.2470	0.2470	0.2470	0.2470	0.2470
Isopar G	4.9985	4.9985	4.9985	4.9985	4.9985	4.9985
Columed MBQ Crestor	0.5785	0.5785	0.5785	0.5785	0.5785	0.5785
L (PGO) - Croda						
Na Oleate	0.0006	0.0029	0.0059	0.0174	0.0174	0.0029
Oleic Acid	0.0006	0.0029	0.0059	0.0174	0.0029	0.0174
BHT	0.1001	0.1001	0.1001	0.1001	0.1001	0.1001
Butane 40	35.0000	35.0000	35.0000	35.0000	35.0000	35.0000
Charge/Mass ($\times 10^{-4}$ C/Kg)	-2.30	-0.89	-0.75	-0.70	-0.74	-1.89
Lewis base value	20.0	17.5	40.0	50.0	60.0	17.5
γ^- (mJ m ⁻²)						
Lewis acid value	2.5	2.0	1.0	1.0	1.0	5.0
γ^+ (mJ m ⁻²)						
Theoretical	2.92	-	5.16	3.16	10.72	-
Conductivity (μ S cm ⁻¹)						
Bulk Conductivity (μ S cm ⁻¹)	8.93	-	16.56	17.86	18.52	-

EXAMPLE 62 to 68

The following Table provides details of compositions in which an aromatic dopant is added to the formulation of Example 51 but the purity of the surfactant was slightly different.

Example No	Aromatic Dopant	Conc ^a	γ^- (mJ m ⁻²)	γ^+ (mJ m ⁻²)	Charge/ Mass (x 10 ⁻⁴ C/kg)
62	resorcinol	1.00*	25.0	2.5	-2.77
63	2-methoxy cinnam-aldehyde	1.00*	22.5	2.5	-2.50
64	arbutin	1.00*	45.0	2.5	-2.27
65	esculetin	1.00*	35.0	2.5	-2.07
66	trans-stilbene	1.00*	20.0	2.5	-1.86
67	4-isoproxy phenol	1.00*	20.0	2.5	-2.86
68	4-acetoxy- 3-methoxy cinnam-aldehyde	0.22**	20.0	2.5	-2.53

* Concentration in columned MBQ Creston L (PGO) - Croda

** Concentration in final formulation

Ingredients and Availability

Oleic acid ¹	:	Technical (Croda Chemicals)
Lauric acid ¹	:	Reagent Grade (BDH)
Palmitic acid ¹	:	Reagent Grade (BDH)
Teric 17A3 ²	:	C ₁₇ alcohol with 3 moles of ethylene oxide (Orica)
Teric 17A2 ²	:	C ₁₇ alcohol with 2 moles of ethylene oxide (Orica)
Teric 12A2 ²	:	C ₁₂ alcohol with 2 moles of ethylene oxide (Orica)
Crill 45 ²	:	Sorbitan trioleate (Orica)
Polyglycerol oleate ²	:	Croda Chemicals
BHT ⁴	:	Butylated hydroxytoluene (Orica)
Norpar 13 ⁵	:	liquid n-paraffin (Exxon)
Bioallethrin ³	:	93% w/w (Agrevo)
Bioresmethrin ³	:	93% w/w (Agrevo)
H46 ⁶	:	16% w/w propane/butane blend (Boral)
H55 ⁶	:	26% w/w propane/butane blend (Boral)
1: Ionic Compound (d)		2: Non-ionic surfactant
3: Insecticide		4: Antioxidant
5: Solvent		6: Propellant

CLAIMS:

1. An electrically neutral composition in the form of a water-in-oil or an oil-in-water emulsion, in which droplets of the emulsion on discharge from an aerosol spray device are imparted with a unipolar electrostatic charge, which composition comprises:
 - (a) at least one propellant in an amount of from 2 to 80% w/w;
 - (b) at least one non-ionic surfactant in an amount of from 0.01 to 10% w/w;
 - (c) optionally one or more oils or solvents, preferably aliphatic, linearly conjugated or aromatic, within the oil phase in an amount of up to 80% w/w.
 - (d) at least one polar or ionic or aromatic or linearly conjugated compound in an amount of from 0.01 to 80% w/w based on the non-ionic surfactant, but which is such that the theoretical conductivity of the emulsion is less than the bulk conductivity of the emulsion; and
 - (e) water.
2. A composition as claimed in claim 1 wherein the difference between the theoretical conductivity of the emulsion and the bulk conductivity of the emulsion is at least $+ 0.5 \mu\text{S cm}^{-1}$.
3. A composition as claimed in claim 2 wherein the difference between the theoretical conductivity of the emulsion and the bulk conductivity of the emulsion is at least $+ 4 \mu\text{S cm}^{-1}$.
4. A composition as claimed in claim 2 wherein the difference between the theoretical conductivity of

the emulsion and the bulk conductivity of the emulsion is at least + 6 μ S cm⁻¹.

5. A composition as claimed in any one of the preceding claims wherein at least 90% by volume of the droplets of the disperse phase within the emulsion have an average diameter of less than 60 μ m.

6. A composition as claimed in claim 5 wherein at least 90% by volume of the droplets of the disperse phase within the emulsion have an average diameter in the range of from 20 to 40 μ m.

7. A composition as claimed in any one of the preceding claims wherein at least one non-ionic surfactant is selected from mono, di and tri sorbitan esters, polyoxyethylene mono, di and tri sorbitan esters; mono and polyglyceryl esters; alkoxylated alcohols; alkoxylated amines; alkoxylated acids; amine oxides; 20 ethoxylated/proxylated block copolymers; alkoxylated alkanolamides; and alkoxylated alkyl phenols.

8. A composition as claimed in claim 7 wherein the ionic surfactant contains at least one alkyl, allyl or substituted phenyl group containing at least one C₆ to C₂₂ carbon chain.

9. A composition as claimed in any one of the preceding claims wherein component (d) is selected from 30 a) alkali metal salts, alkaline earth metal salts, ammonium salts, amine salts or amino alcohol salts of one or more of the following compounds: alkyl sulphates, alkyl ether sulphates, alkylamidoether sulphates, alkylarylpolyether sulphates, monoglyceride

sulphates, polyglyceride sulphates, alkyl sulphonates, alkylamine sulphonates, alkyl-aryl sulphonates, olefin sulphonates, paraffin sulphonates, alkyl sulpho-
succinates, alkylether sulphosuccinates, alkylamide
5 sulphosuccinates, alkyl sulphocinnamates, alkyl sulphoacetates, alkyl phosphates, alkylether phosphates, acyl sarcosinates, acyl isothionates and N-acyl taurates;

b) alkyl amidopropylbetaines, alkylamido-
betaines, alkylamidosulphobetaines, alkylbetaines,
10 aminimides, quaternary ammonium compounds and quaternary
phosphonium compounds;

c) carboxylic acids, carboxylic acid salts,
esters, ketones, aldehydes, amides or amines of
carboxylic acids containing from 6 to 30 carbon atoms;

15 d) diethyl orthophthalate, methylphenylcarbonyl
acetate, α -methyl ionone, 4-hydroxy 3-methoxy-
benzaldehyde, phenylethyl alcohol, dipropylene glycol,
styryl acetate, n-butyl benzoate, isopropyl 4-
hydroxybenzoate, isobutyl acetophenone, isopropyl
20 acetophenone, nicotinic acid, benzoic acid, 2-naphthol,
neopentyl benzene, naphthalene, toluene, fullerene,
tannic acid, t-butylacetophenone, isopropylcinnamate,
resorcinol, 4-methoxycinnamaldehyde, arbutin, 4-acetoxy-
3-methoxycinnamaldehyde, 4-isopropylphenol, trans-
25 stilbene, esculetin, p-chloro-m-xlenol, chloro-o-cresol,
triclosan, norfenefrine, norepinephrine, hexyl-
resorcinol, limonene, methylphenylcarbonyl acetate and p-
tert-butyl- α -methylhydrocinnamic aldehyde.

30 10. A composition as claimed in any one of the
preceding claims wherein component (d) is present in the
composition in an amount of from 0.01 to 30% w/w,
preferably 0.01 to 10% w/w based on the weight of
component (b).

11. A composition as claimed in any one of the preceding claims wherein the droplets formed on discharge from an aerosol spray device have a charge to mass ratio of at least $+/- 1 \times 10^{-4}$ C/kg, preferably at least $+/- 2 \times 10^{-4}$ C/kg.

12. A composition as claimed in any one of the preceding claims which is an insecticidal composition which includes one or more insecticides therein in an 10 amount of from 0.001 to 5% w/w.

13. A method of enhancing the unipolar charge which is imparted to droplets of an emulsion on discharge from an aerosol spray device in which the droplets are 15 formed from an oil-in-water or a water-in-oil emulsion composition which comprises:

(a) at least one propellant in an amount of from 2 to 80% w/w;

20 (b) at least one non-ionic surfactant in an amount of from 0.01 to 10% w/w;

(c) optionally one or more oils or solvents, preferably aliphatic, linearly conjugated or aromatic, within the oil phase in an amount of up to 80% w/w. preferably up to 40% w/w;

25 (d) at least one polar or ionic or aromatic or linearly conjugated compound in an amount of from 0.1 to 80% w/w based on the non-ionic surfactant, but which is such that the theoretical conductivity of the emulsion is less than the bulk conductivity of the emulsion; and

30 (e) water.

14. The use of a non-ionic surfactant and at least one polar or ionic or aromatic or conjugated compound in an amount of from 0.01 to 80% w/w based on the non-ionic

surfactant to enhance the electrostatic charge imparted to droplets of a composition in the form of a water-in-oil or an oil-in-water emulsion on discharge from an aerosol spray device, which composition includes:

5 (a) at least one propellant in an amount of from 2 to 80% w/w;

(b) optionally one or more oils or solvents, preferably aliphatic, linearly conjugated or aromatic, within the oil phase in an amount of up to 80% w/w,
10 preferably up to 40% w/w; and

(c) water;
and the amount of the polar or ionic or aromatic or conjugated compound being such that the theoretical conductivity of the emulsion is less than the bulk
15 conductivity of the emulsion.

15. An aerosol spray device which contains an electrically neutral composition in the form of a water-in-oil emulsion, an oil-in-water emulsion or a single
20 phase composition, in which droplets of the composition on discharge from the aerosol spray device are imparted with a unipolar electrostatic charge, wherein the formulation of the composition and the material of the portion of the aerosol spray device with which the liquid
25 comes into contact on spraying are selected such that

i) the difference between the Lewis base component of the liquid and the Lewis base component of the material with which the liquid comes into contact on spraying is at least $\pm 2\text{mJ m}^{-2}$;
30 and/or:

ii) the difference between the Lewis acid component of the liquid and the Lewis acid component of the material with which the

liquid comes into contact on spraying is at least $\pm 0.5 \text{ mJ m}^{-2}$.

16. An aerosol spray device as claimed in claim 15 wherein the difference in i) is at least $\pm 5 \text{ mJ m}^{-2}$ and/or 5 the difference in ii) is at least $\pm 1 \text{ mJ m}^{-2}$.

17. An aerosol spray device as claimed in claim 15 or 16 wherein the difference i) is at least $\pm 15 \text{ mJ m}^{-2}$ and/or the difference in ii) is at least $\pm 2 \text{ mJ m}^{-2}$.

10

18. An aerosol spray device as claimed in any one of claims 15 to 17 wherein the composition contained therein is a composition as claimed in any one of claims 1 to 14.

15

20

25

30

Fig.1a.

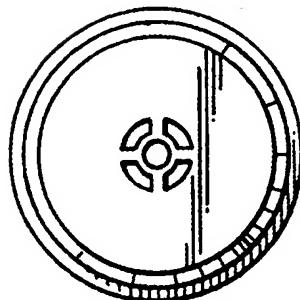


Fig.1b.

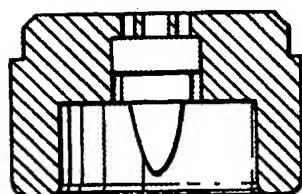


Fig.1c.

